



Europäisches
Patentamt

European
Patent Office

Office européen
des brevets

10/519939
PCT/EP03/07327

29 DEC 2004

REC'D 28 AUG 2003

WIPO PCT

Bescheinigung

Certificate

Attestation

Die angehefteten Unterla-
gen stimmen mit der
ursprünglich eingereichten
Fassung der auf dem näch-
sten Blatt bezeichneten
europäischen Patentanmel-
dung überein.

The attached documents
are exact copies of the
European patent application
described on the following
page, as originally filed.

Les documents fixés à
cette attestation sont
conformes à la version
initialement déposée de
la demande de brevet
européen spécifiée à la
page suivante.

Patentanmeldung Nr. Patent application No. Demande de brevet n°

02077756.1

**PRIORITY
DOCUMENT**
SUBMITTED OR TRANSMITTED IN
COMPLIANCE WITH RULE 17.1(a) OR (b)

Der Präsident des Europäischen Patentamts;
Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets
p.o.

R C van Dijk

BEST AVAILABLE COPY



Anmeldung Nr:
Application no.: 02077756.1
Demande no:

Anmeldetag:
Date of filing: 10.07.02
Date de dépôt:

Anmelder/Applicant(s)/Demandeur(s):

Akzo Nobel N.V.
Velperweg 76
6824 BM Arnhem
PAYS-BAS

Bezeichnung der Erfindung/Title of the invention/Titre de l'invention:
(Falls die Bezeichnung der Erfindung nicht angegeben ist, siehe Beschreibung.
If no title is shown please refer to the description.
Si aucun titre n'est indiqué se référer à la description.)

Depolymerization of water soluble polysaccharides

In Anspruch genommene Priorität(en) / Priority(ies) claimed /Priorité(s)
revendiquée(s)
Staat/Tag/Aktenzeichen/State/Date/File no./Pays/Date/Numéro de dépôt:

Internationale Patentklassifikation/International Patent Classification/
Classification internationale des brevets:

C08B/

Am Anmeldetag benannte Vertragsstaaten/Contracting states designated at date of
filing/Etats contractants désignées lors du dépôt:

AT BE BG CH CY CZ DE DK EE ES FI FR GB GR IE IT LI LU MC NL PT SE SK TR

ACM 2948 PDEP

1

DEPOLYMERIZATION OF WATER SOLUBLE POLYSACCHARIDES

The present invention relates to a process for reducing the molecular weight of a water soluble polysaccharide or polysaccharide ether and to a solid composition comprising a water soluble polysaccharide ether.

Low molecular weight, water soluble polysaccharides, in particular water soluble polysaccharide ethers such as sodium carboxymethyl cellulose, often referred to as carboxymethyl cellulose, are used in various applications, for example, in the paper making industry and in froth flotation for mineral separation. For paper applications, especially in the tissue industry, there is a need for low viscous carboxymethyl cellulose-containing formulations having a high solids content. Such formulations can only be prepared from low molecular weight water soluble polysaccharides or polysaccharide ethers. Froth flotation is the commonly used process for mineral upgrading separating precious metals from useless gangue minerals. Low molecular weight polysaccharides and polysaccharide ethers, such as low viscous carboxymethyl cellulose, are considered to be more efficient in depressing said gangue minerals than high molecular weight polysaccharides.

Low molecular weight polysaccharides may be obtained from higher molecular weight polysaccharides by reducing the molecular weight. Low molecular weight water soluble polysaccharide ethers may be obtained either by appropriately choosing the starting material for the preparation of the polysaccharide ether or they may be produced from higher molecular weight polysaccharides or polysaccharide ethers by reducing the molecular weight during or after their synthesis.

In the prior art, an aqueous hydrogen peroxide solution is generally used for reducing the molecular weight of polysaccharides and polysaccharide ethers. For example, US 6,054,511 discloses a process for producing a high solids, low

ACM 2948 PDEP

2

viscosity aqueous polysaccharide composition comprising stepwise or continuously reacting a polysaccharide or polysaccharide ether with hydrogen peroxide to produce aqueous compositions with a solids content of greater than 5 wt% and a viscosity at 25°C of below 9,500 mPa.s. Preferably, a 30-50%
5 aqueous hydrogen peroxide solution is used for the depolymerization of the polysaccharide or polysaccharide ether.

EP 0 136 722 discloses a process for preparing a carboxymethyl ethyl cellulose which is suitable for use in enteric coating. The process comprises
10 depolymerizing carboxymethyl ethyl cellulose by dissolving carboxymethyl ethyl cellulose in an aqueous solution of a basic compound and adding a peroxide to the solution. After depolymerization, the resulting basic solution is neutralized with an acid. Preferable examples of the basic compound are ammonia, water-soluble amines, and alkali metal hydroxides. The preferred water soluble
15 peroxide is hydrogen peroxide. The depolymerization is carried out in the presence of a basic compound in order to reduce the number of ester linkages, i.e. to reduce the degree of esterification.

A disadvantage of using hydrogen peroxide is that the depolymerization of the
20 polysaccharide or polysaccharide ether takes several hours, typically about 4 to 7 hours in the examples of US 6,054,511. In the examples of EP 0 136 722, depolymerization reaction times of about 5 to 6 hours are reported. A further drawback is that any remaining hydrogen peroxide must be destroyed before the polysaccharide or polysaccharide ether is recovered and this presents a
25 safety problem. Also, the hydrogen peroxide is only available in the form of an aqueous solution and this presents a handling, storage, and transporting problem.

Another disadvantage of hydrogen peroxide is that its use does not always lead to a degree of depolymerization which is desired by the paper making industry,

ACM 2948 PDEP

3

especially when the polysaccharide ether is manufactured in a so-called dry process.

US 5,708,162 discloses a process for the preparation of a low molecular weight polysaccharide ether comprising initially introducing a relatively high molecular weight polysaccharide ether in suspension, e.g. a slurry, adding a perborate, and carrying out an oxidative degradation in an alkaline medium at a temperature of between 25°C and 90°C. Typically, the polysaccharide ether starting materials, in particular cellulose ethers, are also prepared in suspension. The depolymerized polysaccharide ether product is isolated in a dry form.

A disadvantage of this process is that the depolymerization takes place in a suspension, typically using isopropanol or a mixture of isopropanol and water. The use of organic solvents such as isopropanol is not desirable and presents a waste and environmental problem. It also increases the volume of the starting material and final product and thus adds to the manufacturing, storage, and transporting costs. Furthermore, the low molecular weight polysaccharide ether suspension that is formed during the process of US 5,708,162 in particular is not suitable for use in the paper making industry which requires aqueous solutions of low viscosity and having a high solids content. It is of course disadvantageous to first isolate and dry the depolymerized polysaccharide ether product and then dissolve it in water.

WO 01/07485 discloses a process for the depolymerization of polysaccharides or polysaccharide derivatives at increased temperatures comprising (i) mixing at least one polysaccharide with a predetermined amount of at least one peroxo compound and (ii) optionally reacting the polysaccharide of the mixture with a derivatization reagent in order to form a polysaccharide derivative. This document further discloses a mixture comprising at least one polysaccharide

ACM 2948 PDEP

4

and at least one peroxo compound. It is mentioned that the disclosed process makes it possible to depolymerize polysaccharides in one step and to be able to simultaneously or subsequently produce derivatives having a desired degree of polymerization. Suitable polysaccharides are starch, cellulose, inulin, chitin, alginic acid, and guar gum. Suitable peroxo compounds are urea hydrogen peroxide (i.e. "Percarbamid" or carbamide peroxide), percarbonate, and perborate. All examples in this document relate to the preparation of low molecular weight cellulose carbamate using urea and urea hydrogen peroxide.

- 10 The depolymerization or depolymerization/derivatization of the polysaccharide in accordance with the process of WO 01/07485 is carried out in a suspension of xylene and this presents several disadvantages as described hereinabove. We found that the use of carbamide peroxide as such does not lead to the desired reduction in viscosity of the polymer within a reasonable period of time and thus is unsuitable for use commercially.

V.N. Kisenko and E.I. Kuryatnikov in the Russian Journal of General Chemistry, Vol. 70, 2000, pp. 1410-1412, describe the kinetics of degradation of water soluble cellulose ethers such as carboxymethyl cellulose, hydroxyethyl cellulose and methyl cellulose under the action of ammonium persulfate. This document does not disclose or suggest the process of the present invention.

The present invention provides a solution to the above-mentioned problems.

- 25 In accordance with the present invention a process for reducing the molecular weight of a water soluble polysaccharide or polysaccharide ether is provided, said process comprising contacting said polysaccharide or polysaccharide ether with a solid, water soluble depolymerization agent containing 0.25-15 mole equivalents of a base per mole of depolymerization agent, said contacting being carried out in the form of an aqueous solution.

ACM 2948 PDEP

5

The present invention further relates to a solid composition consisting of a water soluble polysaccharide ether and a solid, water soluble depolymerization agent containing 0.25-15 mole equivalents of a base per mole of depolymerization agent.

Apart from avoiding the afore-mentioned drawbacks, the present invention provides the industry with a composition having the desired low viscosity and high solids content upon dissolution in water. The invention process can be carried out in one step, within an acceptable time period, with nearly complete consumption of the depolymerization agent, and making use of readily available higher molecular weight polysaccharide ethers.

In accordance with the present invention, preferably a solid, water soluble depolymerization agent containing 0.25-10, more preferably 0.25-5, even more preferably 0.5-2 mole equivalents of a base per mole of depolymerization agent, is used.

Suitable examples of solid, water soluble depolymerization agents have been described by N. Steiner and W. Eul on Peroxides and Peroxide Compounds, Inorganic Peroxides in Kirk-Othmer Encyclopedia of Chemical Technology, John Wiley & Sons, Inc. 2001 (online posting date of July 13, 2001), in particular Chapter 3 on Group 13 (IIIB) peroxides, Chapter 6 on Group 16 (VIB) peroxides, and Chapter 8 on peroxohydrates, and by J. Sanchez and T.N. Myers on Peroxides and Peroxide Compounds, Organic Peroxides in Kirk-Othmer Encyclopedia of Chemical Technology, John Wiley & Sons, Inc. 1996 (online posting date of December 4, 2000), in particular Chapter 6 on peroxyacids.

Suitable examples of solid, water soluble depolymerization agents for use in accordance with the present invention include sodium percarbonate, sodium

perborate, carbamide peroxide in combination with a base, sodium persulfate in combination with a base, 3-chloroperoxybenzoic acid (m-CPBA) in combination with a base, and mixtures thereof. For example, sodium percarbonate contains approx. 0.7 mole equivalents of sodium carbonate per mole of sodium percarbonate. Any base may be used in accordance with the present invention. Suitable examples include sodium hydroxide and sodium carbonate. A preferred base is sodium carbonate.

In accordance with the present invention, preferably sodium percarbonate or sodium perborate is used. Most preferably, sodium percarbonate is used.

The above-mentioned examples of suitable solid, water soluble depolymerization agents that can be used in accordance with the present invention are commercially available and are relatively cheap materials. This has the advantage that solid mixtures of the water soluble polysaccharide or polysaccharide ether and the solid, water soluble depolymerization agent can be prepared. Any desired final viscosity of the aqueous polysaccharide or polysaccharide ether solution can be obtained by determining the required amount of depolymerization agent in the solid composition using routine experimentation.

It was found that said solid compositions can be easily stored, transported, and handled. The depolymerization of the polymer can be initiated by addition of the solid composition of the polysaccharide ether and the depolymerization agent to water, typically tap water, and stirring for an appropriate period of time, if desired at an elevated temperature. The polysaccharide ether and depolymerization agent can also be added simultaneously, but separately to the water.

When the polysaccharide or polysaccharide ether is added to an aqueous solution of the depolymerization agent, the depolymerization agent may

ACM 2948 PDEP

7.

decompose before it is able to depolymerize the polymer and this reduces the efficiency of the depolymerization reaction. When the polysaccharide or polysaccharide ether is first dissolved in water and then the depolymerization agent is added, an intermediate aqueous solution of high viscosity may be formed. If these problems occur, which can easily be determined by one of ordinary skill in the art, these ways of carrying out the invention process preferably should not be used.

Preferably, the polysaccharide or polysaccharide ether and the depolymerization agent are added simultaneously to the water, either separately or in the form of a solid composition of the polysaccharide or polysaccharide ether and the depolymerization agent. The depolymerization of the polysaccharide or polysaccharide ether takes place after and during dissolution of the polymer in water.

An advantage of the use of the solid composition consisting of a water soluble polysaccharide ether and a solid, water soluble depolymerization agent in accordance with the present invention is that the depolymerization of the polysaccharide ether can be carried out, for example, by a supplier of the paper making industry or by the paper making industry itself just before the low viscous, high solids aqueous solution is needed.

The depolymerization in accordance with the present invention can be carried out using conventional equipment, for example a stirred all glass or stainless steel reactor.

The invention process can be carried out over a wide temperature range - a practical range being 25 to 95°C - typically by stirring at a selected temperature for a period of time until the final or desired viscosity is obtained. In the industry, pumps that are used for dosing chemicals such as carboxymethyl cellulose

typically can handle viscosities up to 1,000 mPa.s (Brookfield LV rheometer, 25°C, 30 rpm). The optimum time, temperature, and stirring conditions can be determined by one of ordinary skill in the art using routine experimentation and the present specification and the Examples described below as guidelines. In
5 the industry, aqueous solutions having viscosities generally of between 1 and 1,000, preferably of between 20 and 600 mPa.s (Brookfield LV rheometer, 25°C, 30 rpm), and having solids contents of between 1 and 40 wt%, preferably 2 to 20 wt%, based on the total weight of the aqueous composition, are used.

10 It is advantageous to carry out the depolymerization in accordance with the present invention at an elevated temperature. A preferred temperature range is 35 to 80°C. A more preferred temperature range is 45 to 70°C. In general, under the conditions that are described in the Examples of this application, a reaction temperature of 45°C or higher results in a depolymerization reaction time of 1
15 hour or less.

The resulting aqueous solution that is obtained in accordance with the process of the present invention is ready for use in the industry and need not undergo any further treatment.

20

In accordance with the present invention, any water soluble polysaccharide or polysaccharide ether can be used. Suitable examples have been described as water soluble gums by J.N. BeMiller on Carbohydrates in Kirk-Othmer Encyclopedia of Chemical Technology, John Wiley & Sons, Inc. 1992 (online
25 posting date of December 4, 2000), in particular in Chapter 5. Either technical or purified grades of these polymers can be used.

Suitable examples of water soluble polysaccharides include guar gum, dextrin, xanthan gum, carrageenan, and gum arabic. Suitable examples of water soluble
30 polysaccharide ethers include the carboxymethyl (CM), hydroxypropyl (HP),

ACM 2948 PDEP

9

hydroxyethyl (HE), ethyl (E), methyl (M), hydrophobically modified (HM), quaternary ammonium (QN), and mixed ether derivatives of cellulose (C), guar (G), and starch (S), such as HEC, HPC, EHEC, CMHEC, HPHEC, MC, MHPC, MHEC, MEHEC, CMC, CMMC, CMG, HEG, HPG, CMHPG, HMCMC, HMHEC, 5 HMHPC, HMEHEC, HMCMEC, HMHPHEC, HMMC, HMMHPC, HMMHEC, HMCMMC, HMG, HMCMG, HMHEG, HMHPG, HMCMPG, QNCMC, and HPS. These water soluble polysaccharides and polysaccharide ethers are known in the art and either are commercially available or can be manufactured using methods known per se in the art. The carboxymethyl derivatives generally 10 are used in the form of an alkali metal salt, usually the sodium salt thereof.

Preferably, a water soluble polysaccharide ether is used in accordance with the present invention. Preferably, the water soluble polysaccharide ether is selected from the group consisting of CMC, HMCMC, HEC, HMHEC, EHEC, and 15 HMEHEC. More preferably, the polysaccharide ether is CMC or carboxymethyl cellulose.

The molecular weight (Mw) of the water soluble polysaccharide or polysaccharide ether to be used in accordance with the present invention can 20 vary over a wide range. The molecular weight typically is in the range of 25,000 to 3,000,000, preferably 25,000 to 500,000, more preferably 50,000 to 250,000 Dalton.

The amount of water soluble polysaccharide or polysaccharide ether to be used 25 in accordance with the present invention can vary over a wide range. It typically depends on the desired solids content of the resulting aqueous solution. Generally, in the industry, a solids content of 1 to 25 wt%, based on the total weight of the aqueous composition, is used. In the paper industry, a solids content of preferably 5 to 15, more preferably 7 to 10 wt% is used.

30

The amount of solid, water soluble depolymerization agent to be used in accordance with the present invention also can vary over a wide range and typically will be determined by the desired final viscosity of the polysaccharide or polysaccharide ether aqueous solution. A practical amount to be used is 0.1
5 to 30, preferably 0.5 to 15, more preferably 2 to 10 wt%, based on the weight of the polysaccharide or polysaccharide ether. The molecular weight of the depolymerized polysaccharide or polysaccharide ether typically is in the range of 10,000 to 250,000 Dalton.

10 It is known to the man skilled in the art that peroxide reactions are catalyzed by impurities like transition metal ions ordinarily present in tap water. If necessary, such impurities may be added in a conventional amount to the invention process. Optionally, any of the known activators, which are mentioned for sodium perborate in US 5,708,162, may be used in a conventional amount in
15 the process of the present invention.

The depolymerization of the polysaccharide or polysaccharide ether can easily be followed by determining the viscosity of the aqueous solution. The amount of depolymerization agent in the reaction mixture can be determined as a function
20 of the time in any conventional way. For example, the amount of hydrogen peroxide can be determined by iodometric titration or by using peroxide test sticks, which are commercially available. The resulting aqueous solution containing the depolymerized polysaccharide or polysaccharide ether can be used either when the final or when the desired viscosity is reached, preferably
25 after all depolymerization agent has reacted away. In this way, an aqueous solution is obtained which is safe and easy to handle.

The present invention is illustrated by the following Examples.

ACM 2948 PDEP

11

Example 1

In a stirred stainless steel reactor, 32.5 g of CMC (Akucell AF 0305, ex Akzo Nobel, having a water content of 7.6% and a viscosity of 4,757 mPa.s for a 6 wt% aqueous solution of said CMC when measured with a Brookfield LV rheometer operating at 10 rpm and 25°C) were dissolved in 467.5 g of tap water at 65°C giving a 6 wt% aqueous CMC solution. To this aqueous solution, 0.60 g of sodium percarbonate (ex Aldrich), i.e. 2 wt% relative to the amount of CMC, was added in one minute under vigorous stirring. Samples were taken after 0.25, 0.5 and 1 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 297, 200 and 212 mPa.s, respectively. The percentage hydrogen peroxide consumed was calculated after iodometric titration to be 96, 100 and 100%, respectively. The Mw dropped from 132,000 Dalton at the start to 82,700 Dalton after 1 h of stirring.

15

Example 2

37.5 g of CMC (Akupure 0310, ex Akzo Nobel, having a water content of 8.8% and a viscosity of 2,843 mPa.s for a 6.9 wt% aqueous solution of said CMC when measured with a Brookfield LV rheometer operating at 10 rpm and 25°C) was added in one minute to 462.5 g of tap water of 50°C in a stirred all glass reactor. Then, 1.875 g of sodium percarbonate, i.e. 5 wt% relative to the amount of CMC, were added in one minute under vigorous stirring. Samples were taken after 0.25 and 0.5 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 388 and 239 mPa.s, respectively. The percentage hydrogen peroxide consumed was calculated after iodometric titration to be 78 and 95%, respectively. Upon depolymerization the Mw dropped from 94,000 to 61,000 Dalton.

20
25Example 3

A dry blend of 32.5 g of CMC (Akucell AF 0305, see Example 1) and 0.60 g of sodium percarbonate was made by mixing with a spatula. The blend was added under vigorous stirring in one minute to 467.5 g of tap water of 65°C in a stainless steel reactor. Samples were taken after 0.25 and 0.5 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 365 and 330 mPa.s, respectively. The percentage hydrogen peroxide consumed was calculated after iodometric titration to be 99 and 100% respectively.

10 Example 4

Similar results as in Example 3 were obtained after complete consumption of the sodium percarbonate when the depolymerization was carried out at 55°C, 50°C or 45°C. The lower the temperature, the slower the consumption of sodium percarbonate. Complete consumption of sodium percarbonate at the reaction temperature of 50°C occurred after 0.5 h.

Example 5

A dry blend of 32.0 g of CMC (Akucell AF 0305, see Example 1, but containing 6.2% of water) and 0.88 g of sodium perborate (ex Aldrich) was made by mixing with a spatula. The blend was added under vigorous stirring in one minute to 468 g of tap water of 50°C in an all glass reactor. Samples were taken after 0.25, 0.5, 1 and 2 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 564, 369, 264 and 202 mPa.s, respectively. The percentage hydrogen peroxide consumed was calculated after iodometric titration to be 58, 75, 90 and 98%, respectively.

Example 6

A dry blend of 40.0 g of a technical grade CMC (Gabrosa PA 386, having a water content of 5.8% and a viscosity of 7,049 mPa.s for a 7.5 wt% aqueous solution of said CMC when measured with a Brookfield LV rheometer operating

ACM 2948 PDEP

13

at 1 rpm and 25°C) and 0.80 g of sodium percarbonate was made by mixing with a spatula. The blend was added under vigorous stirring in one minute to 460.0 g of tap water of 67°C in a stainless steel reactor. Samples were taken after 0.25 and 0.5 h of stirring. The viscosity (measured with a Brookfield LV
6 rheometer operating at 30 rpm and 25°C) of these samples was 251 and 233 mPa.s, respectively. The percentage hydrogen peroxide consumed was determined by using peroxide test sticks (Quantofic® Peroxide 25) and was found to be 99 and 100%, respectively. A reduction in Mw from 165,800 Dalton to 63,700 Dalton was observed.

10

Example 7

A dry blend of 32.0 g of CMC (Akucell AF 0305, see Example 1, but containing 6.2% of water), 1.37 g of sodium persulfate (ex Aldrich) and 0.40 gram of sodium carbonate (ex Aldrich) was made by mixing with a spatula. The blend
15 was added under vigorous stirring in one minute to 468.0 g of tap water of 50°C in a stirred all glass reactor. Samples were taken after 0.25, 0.5 and 1.0 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 563, 178 and 63 mPa.s, respectively.

20 Example 8

32.0 g of CMC (Akucell AF 0305, see Example 1, but containing 6.2% of water) was added in one minute to 468.0 g of tap water of 60°C in a stirred all glass reactor. Immediately after the addition of the CMC, 1.40 g of 3-chloroperoxybenzoic acid (ex Akzo Nobel, 71% active content) and 0.40 g of
25 sodium carbonate were added in one minute under vigorous stirring. Samples were taken after 0.25, 0.5 and 1.0 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 495, 295 and 205 mPa.s, respectively. The percentage peroxide consumed was calculated after iodometric titration to be 78, 91 and 97%, respectively. Upon

depolymerization a decrease in Mw was observed from 126,400 to 80,500 Dalton.

Example 9

- 5 In a stirred all glass reactor, 32.2 g of EHEC (Bermocoll E270, ex Akzo Nobel, having a water content of 6.8% and a viscosity of 104,000 mPa.s for a 6 wt% aqueous solution of said EHEC when measured with a Brookfield LV rheometer operating at 0.5 rpm and 25°C) were dissolved in 467.8 g of tap water at 50°C giving a 6 wt% aqueous EHEC solution. To this aqueous solution, 1.50 g of
- 10 sodium percarbonate (ex Aldrich), i.e. 5 wt% relative to the amount of EHEC, was added in one minute under vigorous stirring. Samples were taken after 0.5 and 1.0 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 10 and 30 rpm, respectively, and 25°C) of these samples was 2,693 and 1,128 mPa.s, respectively. The percentage hydrogen peroxide
- 15 consumed was calculated after iodometric titration to be 95 and 98%, respectively.

Example 10

- 20 In a stirred all glass reactor, 21.5 g of EHEC (Bermocoll E320G, ex Akzo Nobel, having a water content of 6.9% and a viscosity of 39,000 mPa.s for a 4 wt% aqueous solution of said EHEC when measured with a Brookfield LV rheometer operating at 1 rpm and 25°C) were dissolved in 4787.5 g of tap water at 55°C giving a 4 wt% aqueous EHEC solution. To this aqueous solution, 1.00 g of
- 25 sodium percarbonate (ex Aldrich), i.e. 5 wt% relative to the amount of EHEC, was added in one minute under vigorous stirring. Samples were taken after 0.25, 0.5 and 1.0 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 460, 329 and 267 mPa.s, respectively. The percentage hydrogen peroxide consumed was calculated after iodometric titration to be 87, 97 and 99%, respectively. A

ACM 2948 PDEP

15

decrease in Mw was observed from 854,700 to 141,800 Dalton after 1.0 h of stirring.

Example 11

5 A dry blend of 11.2 g of guar gum (ex Dinesh Enterprises, having a water content of 10.3% and a viscosity of 350,000 mPa.s for a 2 wt% aqueous solution of said guar gum when measured with a Brookfield LV rheometer operating at 1 rpm and 20°C) and 0.50 g of sodium percarbonate was made by mixing with a spatula. The blend was added under vigorous stirring in one
10 minute to 488.9 g of tap water of 70°C in a stirred all glass reactor. Samples were taken after 1.0 and 2.0 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at 30 rpm and 25°C) of these samples was 301 and 158 mPa.s, respectively. The percentage hydrogen peroxide consumed was calculated after iodometric titration to be 80 and 95%,
15 respectively. Upon depolymerization a decrease in Mw was observed from 1,914,000 to 180,200 Dalton.

Comparative Example A

32.0 g of CMC (Akucell AF 0305, see Example 1, but containing 6.2% of water)
20 was added in one minute to 468.0 g of tap water of 60°C in a stirred all glass reactor. Immediately after the addition of the CMC, 1.40 g of 3-chloroperoxybenzoic acid (ex Akzo Nobel, 71% active content) was added in one minute under vigorous stirring. Samples were taken after 0.5, 1.0, and 2.0 h of stirring. The viscosity (measured with a Brookfield LV rheometer operating at
25 30 rpm and 25°C) of these samples was 859, 592 and 388 mPa.s, respectively. The percentage peroxide consumed was calculated after iodometric titration to be 45, 64 and 80%, respectively.

ACM 2948 PDEP

16

CLAIMS

1. A process for reducing the molecular weight of a water soluble polysaccharide or polysaccharide ether comprising contacting said
5 polysaccharide or polysaccharide ether with a solid, water soluble depolymerization agent containing 0.25-15 mole equivalents of a base per mole of depolymerization agent, said contacting being carried out in the form of an aqueous solution.
- 10 2. A process according to claim 1, characterized in that the polysaccharide or polysaccharide ether and the depolymerization agent are added simultaneously to water.
- 15 3. A process according to claim 1, characterized in that a solid composition consisting of the polysaccharide or polysaccharide ether and the depolymerization agent is added to water.
- 20 4. A process according to any one of claims 1-3, characterized in that the depolymerization agent is selected from the group consisting of sodium percarbonate, sodium perborate, carbamide peroxide in combination with a base, sodium persulfate in combination with a base, 3-chloroperoxybenzoic acid (m-CPBA) in combination with a base, and mixtures thereof.
- 25 5. A process according to any one of claims 1-3, characterized in that the base is sodium hydroxide or sodium carbonate.
6. A process according to claim 4, characterized in that the depolymerization agent is sodium percarbonate or sodium perborate.

ACM 2948 PDEP

17

7. A process according to any one of claims 1-6, characterized in that the water soluble polysaccharide ether is selected from the group consisting of carboxymethyl cellulose, hydrophobically modified carboxymethyl cellulose, hydroxyethyl cellulose, hydrophobically modified hydroxyethyl cellulose, ethyl hydroxyethyl cellulose, and hydrophobically modified ethyl hydroxyethyl cellulose.
8. A solid composition consisting of a water soluble polysaccharide ether and a solid, water soluble depolymerization agent containing 0.25-15 mole equivalents of a base per mole of depolymerization agent.
9. A composition according to claim 8, characterized in that the depolymerization agent is selected from the group consisting of sodium percarbonate, sodium perborate, carbamide peroxide in combination with a base, sodium persulfate in combination with a base, 3-chloroperoxybenzoic acid (m-CPBA) in combination with a base, and mixtures thereof.
- 10.A composition according to claim 9, characterized in that the depolymerization agent is sodium percarbonate or sodium perborate.
- 11.A composition according to any one of claims 8-10, characterized in that the water soluble polysaccharide ether is selected from the group consisting of carboxymethyl cellulose, hydrophobically modified carboxymethyl cellulose, hydroxyethyl cellulose, hydrophobically modified hydroxyethyl cellulose, ethyl hydroxyethyl cellulose, and hydrophobically modified ethyl hydroxyethyl cellulose.
- 12.A composition according to any one of claims 8-11 consisting of carboxymethyl cellulose and sodium percarbonate.

ACM 2948 PDEP

18

ABSTRACT

The invention relates to a process for reducing the molecular weight of a water soluble polysaccharide or polysaccharide ether comprising contacting said
5 polysaccharide or polysaccharide ether with a solid, water soluble depolymerization agent containing 0.25-15 mole equivalents of a base per mole of depolymerization agent, said contacting being carried out in the form of an aqueous solution. The invention also relates to a solid composition consisting of a water soluble polysaccharide ether and a solid, water soluble
10 depolymerization agent containing 0.25-15 mole equivalents of a base per mole of depolymerization agent. Preferably, the depolymerization agent is selected from the group consisting of sodium percarbonate, sodium perborate, carbamide peroxide in combination with a base, sodium persulfate in combination with a base, 3-chloroperoxybenzoic acid in combination with a
15 base, and mixtures thereof. More preferably, the depolymerization agent is sodium percarbonate or sodium perborate. Preferably, the water soluble polysaccharide ether is carboxymethyl cellulose. The present invention provides a process for preparing a high solids, low viscous aqueous solution of a water soluble polysaccharide or polysaccharide ether within a short reaction time in
20 which the depolymerization agent is almost entirely consumed.

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☒ FADED TEXT OR DRAWING
- ☐ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☒ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☒ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.